PROGRESS IN TECHNOLOGY DEVELOPMENT FOR CONVERSION OF ^{99}Mo PRODUCTION FROM HEU TO LEU TARGETS - DEVELOPMEMT OF NEW SEPARATION SCHEMES, AND NEW PROCESSING EQUIPMENT.

Allen J. Bakel, Dominique C. Stepinski, Artem Guelis, Andrew Hebden,
Argentina Leyva, Lohman Hafenrichter, Pauline Gentner, and George F. Vandegrift

Argonne National Laboratory
Chemical Engineering Division
Argonne, IL USA

ABSTRACT

Currently, most of the world's supply of ^{99}Mo is produced from the fissioning ^{235}U in high-enriched uranium (HEU) targets. Conversion of these targets to low-enriched uranium (LEU) would ease worldwide concern over the use and transport of this weapons-grade material. This paper documents our progress in three development areas: (1) separation schemes for ^{99}Mo recovery from both nitrate and sulfate solution-reactor fuels, (2) a small-foot-print dissolver for the LEU-modified Cintichem process, and (3) a dissolver and Mo-recovery system for the low volumes required for alkaline processing of LEU-foil targets.

Introduction

Technecium-99m is the most commonly used radioisotope in nuclear medicine[1]. It is produced from the decay of its parent ^{99}Mo. Nearly all of the ^{99}Mo is produced in research, test or isotope production reactors by irradiation of highly enriched uranium (HEU) targets. The use of HEU causes worldwide concerns over the use and transport of this weapon grade material and generation of larger than necessary radioactive waste inventories[2]. In the United States, additional concerns are raised by the vulnerability of the supply since all of the demand for this important medical isotope is met by foreign producers[3].

Production of ^{99}Mo utilizing LEU instead of HEU fuel eliminates the concerns over proliferation, criticality and transportation of the weapons-grade material. However substitution of LEU for HEU also poses several challenges[3]. To yield equivalent amount of ^{99}Mo, use of LEU requires approximately five times more uranium than HEU to produce the same amount of ^{99}Mo. Consequently, substituting LEU for HEU requires changes in chemical processing to efficiently separate ^{99}Mo from high concentrations of uranium while obtaining a product of equal purity and modifying the processes as little as possible to limit economic disadvantages[1]. This paper discusses methods used for the separation of ^{99}Mo from aqueous acidic media and summarizes our progress on investigating potential new sorbents for recovery of molybdenum from high concentrations of uranium in acidic nitrate or sulfate media.

Recovery of ^{99}Mo from acidic uranyl sulfate and nitrate solutions

Development of a homogeneous reactor for production of ^{99}Mo utilizing low enriched uranium (LEU) for production of ^{99}Mo is underway in the United States and elsewhere. There are many advantages to using a homogeneous reactor for ^{99}Mo production[4]. Aqueous solution reactors are considered to be extremely safe due to a low power coefficient.[5-7] Such reactors can operate under 1/100th of the power and generate 1/100th of the radioactive waste per Curie of ^{99}Mo produced than using conventional target technologies. In a homogeneous reactor, fuel material is dissolved in aqueous media and ^{99}Mo can be periodically separated from the aqueous fuel solution by passing the fuel solution through a Mo-selective column. Thus, use of aqueous fuel eliminates the time and the associated costs of target production and dissolution. The reactor can operate for about 10-20 years before the fuel solution needs replacement and long lived fission products are removed, provided that ^{235}U which has produced ^{99}Mo is regularly replaced and the high cross section neutron poisons are continuously removed during the processing step.[5-7] Nitrate and sulfate media are considered most viable for a homogeneous reactor fuel solution due to considerations of chemical and nuclear stability[4] and the efficiency of fission product separation.

Alumina is the most widely used sorbent for separation of ^{99}Mo from uranium and fission products, its use has been described in various works.[8-10] The chemical process used to extract Curie quantities of ^{99}Mo from irradiated uranium was first developed by Brookhaven National Laboratory.[9] The Brookhaven process, which is the basis for the process used by MDS Nordion, consists of dissolving an irradiated uranium target in nitric acid and passing it through an alumina column which selectively absorbs ^{99}Mo. The alumina column is then washed; ^{99}Mo is stripped from the column with an alkali solution and the bulk ^{99}Mo can undergo a series of purification steps for the preparation of a ^{99m}Tc generator.

Betenekov *et. al.* patented a method of separation of ^{99}Mo from 80 g/L uranyl sulfate solutions at pH=1, from a homogeneous reactor.[11] The Thermoxid sorbents, Radsorb (TiO_2; 75 mol %, SnO_2; 25 mol %)and Isosorb (TiO_2; 95 mol %, ZrO_2; 5 mol %) The sorbents are in a form of pellets 0.1 to 2 mm in size, having a specific surface area of 100 to 350 m^2/g.

Ponomarev-Stepnoy *et. al.* patented a method for production and extraction of ^{99}Mo generated in a uranyl sulfate (pH~1) based homogeneous reactor, which is operated at powers ranging from 20 kW to 100 kW.[12, 13] The sorbent material used to separate ^{99}Mo is a solid polymer composed of a composite ether of a maleic anhydride copolymer and α-benzoin-oxime (ABO). Molybdenum is extracted from the solution with at least 90 % percent efficiency. Less than 2% of other fission products are extracted and less than 0.01% of the uranium is absorbed by the sorbent. This report does not discuss stability of the polymer material to the incurred radiation and acid conditions and does not discuss sorbent regeneration and reuse conditions.

Koehly *et. al.* describe a method for separation of molybdenum from a uranium-aluminium alloy target dissolved in sulfuric acid.[14] The extraction of ^{99}Mo from a sulfuric acid solution of acidity close to 0.1 N is carried out using a column lined with silica gel charged with solution containing 0.05 M of tributylaceto hydroxamic acid in mesitylene. This work does not discuss the stability of extractant and organic solvent to the incurred radiation and acid conditions and does not discuss separation factors of ^{99}Mo from uranium.

Another method that should be mentioned here is separation of ^{99}Mo from uranium fuel solution by selective precipitation with ABO. Variations of this method have been described in various works and involve precipitation from both nitrate and sulfate media.[15-18] Following precipitation, molybdenum is filtered and dissolved and undergoes a series of purification steps.

Experimental

Materials. ^{99}Mo was obtained by stripping a ^{99m}Tc generator (Bristol-Myers Squibb, North Billerica, MA) with 1.0 M NH_4OH, bringing the eluate to dryness and redissolving in 0.1 M HNO_3. The depleted $UO_2(NO_3)_2 \cdot 6H_2O$ was obtained from ANL's stocks and used without further purification. UO_2SO_4 was prepared by dissolving UNH in sulfuric acid and bringing the solution to dryness three times. The Thermoxid sorbents, Radsorb (TiO_2; 75 mol %, SnO_2; 25 mol %)and Isosorb (TiO_2; 95 mol %, ZrO_2; 5 mol %), available as spherical solids of approximately 0.7-1.0 mm and 0.2-0.4 mm in diameter, were used as received. The polyzirconium chloride sorbent (PZC, or PolyZircomium Compound) was received from Kaken Co., Ltd. and was used as received. The alumina was obtained as Acid Alumina AG4, 100-200 mesh (Bio-Rad Laboratories) and was used as received. All other reagents were analytical grade and were used without further purification.

Batch uptake measurements. The sorption of metal ions by the inorganic ion-exchange sorbents from aqueous solutions was measured by contacting a 1 mL volume of a tracer-spiked aqueous solution of appropriate concentration with a known weight of sorbent following procedures described previously.[16-17] A one-hour mixing time was used for the uptake distribution measurements. After contact, the solution was withdrawn and filtered using a syringe fitted with a 0.2 µm pore size, Anotop 25 Inorganic Membrane Filter (Whatman Scientific). Blank experiments indicate that the filter does not uptake ^{99}Mo. Duplicate experiments indicate that the reproducibility of the measurements was generally within 5%; however, the uncertainty interval may be higher for the highest and lowest K_d values. Good activity balance was observed for all experiments.

Counting of aqueous samples was performed on a Minaxi Autogamma counter (gamma emitters) or a Packard model liquid scintillation counter. ^{99}Mo was quantified by measurement of its 739 KeV and 778 KeV γ-rays. The activity of ^{99}Mo in each sample was corrected for decay.

The extent of radionuclide uptake was expressed in terms of a distribution coefficient, K_d, defined as follows:

$$K_d = \left(\frac{A_o - A_s}{W}\right) \Big/ \frac{A_s}{V}$$

Here, A_o and A_s represent the aqueous phase activity (cpm) before and after equilibration, respectively; W is the dry weight of the sorbent (g) and V is the volume of the aqueous phase (mL). The amount of sorbent used was generally kept at 10 (± 1) mg in order to leave a measurable activity in the aqueous phase, this amount of sorbent represents a large excess relative to the amount of radionuclide present.

Uptake kinetics measurements. The rate of radionuclide uptake on the sorbents was measured by contacting a series of equal weight (10 ± 1 mg) samples of sorbents with a tracer-spiked aqueous solution, as described previously.[18] Time zero was taken as the time at which a known volume of the tracer-spiked solution was introduced into the vials containing the sorbent and stirring commenced. At various time intervals, the aqueous phase was withdrawn from a test tube (thus establishing t_{final}) and filtered.

Results and Discussion-Evaluation of Radsorb, Isosorb (Thermoxid) and PZC as potential sorbents for separation of ^{99}Mo from a homogeneous reactor fuel solution

Kinetics of molybdenum uptake. Figure 1 shows the kinetics of uptake of trace levels of ^{99}Mo by the two Thermoxid sorbents (Radsord, Isosorb) and PZC sorbents from 0.1 M HNO_3

solution. In all cases, the equilibrium sorption achieved within 60 minutes is far greater than the minimum required for satisfactory performance in a packed column. To yield a suitable retention in a column mode, a K_d of only few hundred is required; therefore, a satisfactory uptake in all cases is achieved in only 15 minutes (Figure 1).*Evaluation of ^{99}Mo uptake from HNO_3 solutions*. The uptake of trace levels of ^{99}Mo by the Thermoxid and PZC sorbents was also evaluated as a function of nitric-acid concentration (Figure 2). It was found that the uptake of ^{99}Mo at nitric acid concentrations of 0.1 M and lower are practically the same for the Thermoxid sorbents and alumina, whereas uptake of ^{99}Mo by the PZC is significantly lower. At nitric acid concentrations higher than 0.1M, the uptake of ^{99}Mo by the Thermoxid and PZC sorbents is practically the same; whereas, uptake by alumina drops off significantly.

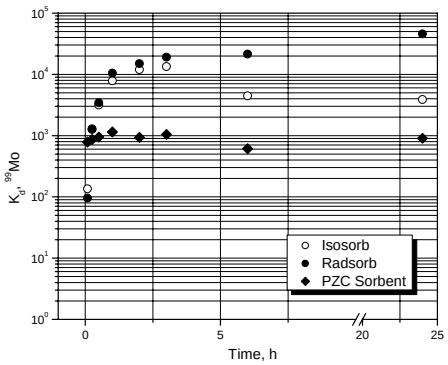

Figure 1. Kinetic of uptake of ^{99}Mo from 0.1 M HNO_3 solutions by the Thermoxid and PZC sorbents.

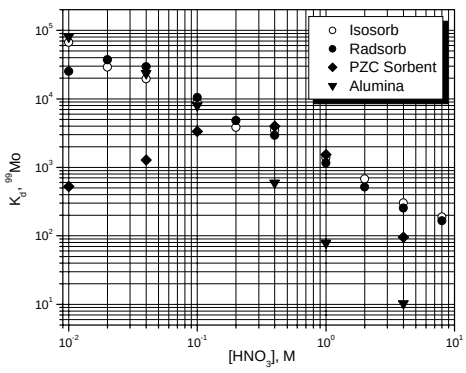

Figure 2. Effect of increasing nitric acid concentrations on the uptake of ^{99}Mo by the Thermoxid sorbents, PZC and Alumina.

Evaluation of ^{99}Mo separation from uranium nitrate solutions. The efficiency of the Thermoxid and PZC sorbent for separation of Mo from solutions containing high concentrations of uranium was evaluated by examining the uptake of trace levels of ^{99}Mo by Isosorb, Radsorb, PZC and alumina sorbent from 1 M HNO_3 solutions of variable uranium concentrations (Figure 3). It was found that the Thermoxid and PZC sorbents all have a very good retention of ^{99}Mo, up to uranium concentrations of 310 g/L. The uptake of ^{99}Mo by Thermoxid and PZC sorbents is over an order of magnitude higher than that observed for Alumina over the entire range of uranium concentrations investigated. The comparison of the Thermoxid sorbents and PZC reveals the ^{99}Mo is equally well extracted up to uranium concentrations of 150 g/L. At uranium concentrations above 150 g/L, the uptake values for ^{99}Mo by both Thermoxid sorbents are higher than those observed for the PZC sorbent. Retention of ^{99}Mo by alumina from uranium solutions is significantly lower than that observed for Thermoxid and PZC sorbents. The K_d values of <50 over the entire range of uranium concentrations indicate low uptake of ^{99}Mo and is inadequate for separation of molybdenum from a homogeneous reactor containing LEU fuel.

Determination of sorbent capacity. The capacity of a sorbent is an important property because it determines the size of the primary separation/purification column. The capacity of the Thermoxid sorbents, Radsor and Isosorb (0.2-0.4 mm particle size) and PZC (polyzirconium chloride) sorbents for Mo from 1 M HNO_3 was tested (Figure 4). The uptake was measured by contacting equal (± 10%) amounts of sorbent with nitric acid solutions of increasing

molybdenum concentration. Under conditions with no uranyl salts are present, it was found that the PZC sorbent has a capacity of ≥5 meq-Mo/g sorbent. The Thermoxid sorbents, Radsorb and Isosorb have a capacity of ≥2.8 and ≥3 meq Mo/g sorbent, respectively. Sorbent capacity, along with Mo uptake from uranium solutions and kinetic data will be used to determine the size of the primary column needed to separate Mo from a volume of LEU solution.

Evaluation of ^{99}Mo uptake from H_2SO_4 solutions. Figure 5 depicts the uptake of trace levels of ^{99}Mo by the Radsorb, Isosorb, PZC and alumina sorbents from sulfuric acid solutions. Sorption of ^{99}Mo from sulfuric acid solutions by the Thermoxid sorbents is significantly higher than by the PZC sorbent. Alumina weakly adsorbs ^{99}Mo from sulfuric acid solutions, the K_d value of 70 in 0.1 M H_2SO_4 implies that alumina may not be appropriate for separation of ^{99}Mo from a homogeneous reactor fuel.

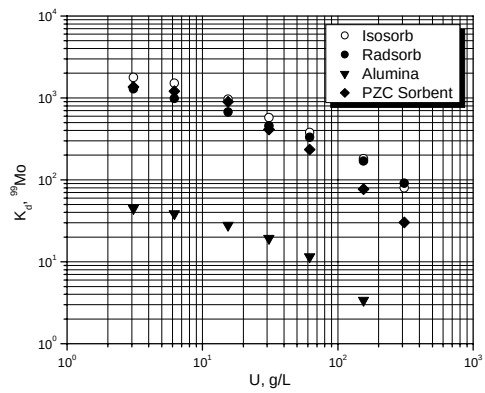

Figure 3. Effect of varying uranium concentrations on the uptake of trace levels of ^{99}Mo from 1 M HNO_3.

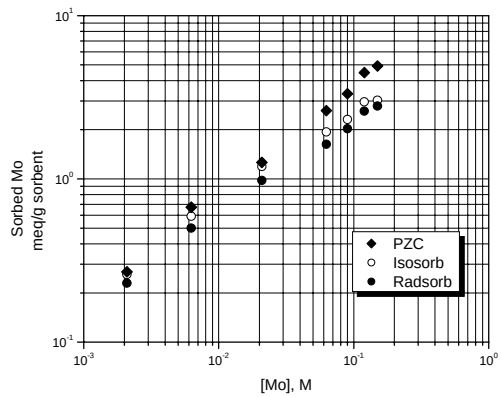

Figure 4. Capacity of PZC and Thermoxid sorbents for uptake of Mo from 1 M HNO_3 solution.

Evaluation of ^{99}Mo separation from uranium sulfate solutions. To identify the most promising sorbents for the extraction of ^{99}Mo from uranyl sulfate solutions, a candidate media for a aqueous homogeneous reactor, uptake of trace levels of ^{99}Mo from solutions of variable uranium sulfate content at pH 1 by PZC and Thermoxid sorbents was evaluated (Figure 6). It was found that PZC sorbent has low retention of ^{99}Mo when contacted with 10-400 g/L uranium solutions and will not be appropriate for separation of Mo from uranium sulfate solutions in a column mode. The Thermoxid sorbents have a higher uptake of Mo (almost 2 orders of magnitude) from uranium sulfate solutions than PZC. The molybdenum distribution ratio values for Radsorb and Isosorb from 350 g/L uranium sulfate solutions at pH 1 are 130 and 150 respectively, which suggest that Mo can be separated from uranium sulfate using these sorbents in a column mode.

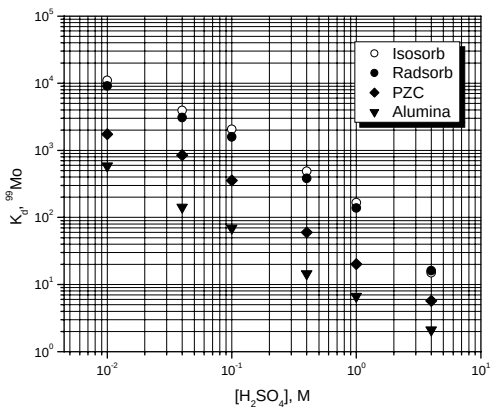

Figure 5. Effect of increasing sulfuric acid concentrations on the uptake of ^{99}Mo by the Thermoxid, PZC and Alumina sorbents.

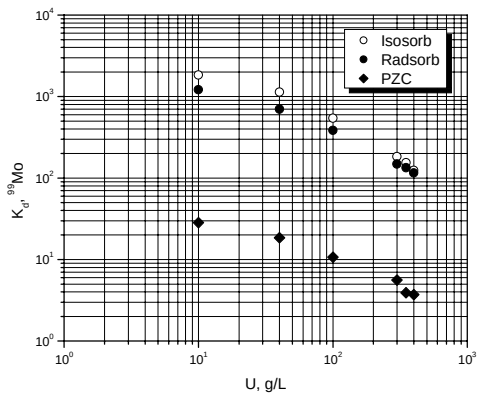

Figure 6. Effect of uranium concentrations on the uptake of of ^{99}Mo from H_2SO_4 solutions at pH 1 by the Thermoxid and PZC sorbents.

Effect of a radiation field on the sorption behaviour of Mo.

High radiation fields are known to affect chemical behaviours in aqueous solutions[5]. The effect of beta and gamma radiation on Mo speciation in acidic media and its specific sorption behavior on Thermoxid anion exchange resin (Radsorb) has been studied. Solutions of nitric and sulfuric acids with and without corresponding uranyl salts were irradiated in the electron beam at Argonne's Van-der-Graaff generator. The effect of radiation on the oxidation state of Mo is expected to be an important factor for Mo sorption. The solutions irradiated contained 10 mg Mo(VI)/L, and 300 g U/L. The initial measured pH was 1.0, and the pH of the solutions did not change during the irradiation. The Kd (Mo) was measured for the sorption of Mo on Radsorb from irradiated and unirradiated solutions approximately four hours after irradiation (Figure 7). The behaviours of both nitrate and sulphate solutions, with and without uranium were evaluated.

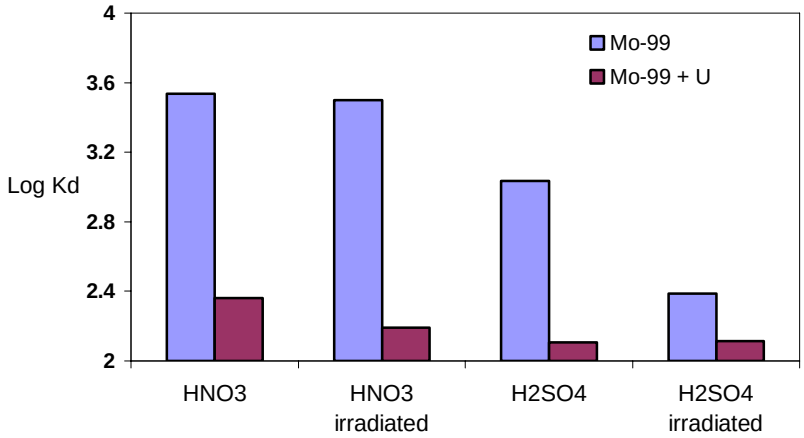

Figure **7**. Measured Kd (Mo) values for the sorption of ^{99}Mo sorption onto the Radsorb in both irradiated and unirradiated solutions. Nitrate and sulphate solutions with and without uranium were used.

The data in Figure 7 show that, after four hours of standing, Mo sorption from nitric acid with no uranium was not affected by irradiation; the K_d from the unirradiated system was 3420, while the K_d from the irradiated system was 3150. As expected (see Figure 2 and 8) the presence of 300g-U/L leads to a decrease in the measured distribution ratios for both irradiated and unirradiated solutions. In addition, the measured K_d is somewhat lower for the irradiated solution (155) than in the unirradiated solution (230).

The data in figure 7 show that the sorption of Mo, in the absence of uranium is less efficient from sulfate solutions than from nitrate solutions. Irradiation caused a decrease of K_d from ~1100 to 240 in the uranium-free sulfate solutions. On the other hand, the K_d values measured for irradiated and unirradiated solutions in the presence of 300 g U/L are identical at about 130.

The decrease of K_d following irradiation in U-free sulfuric acid solutions can be explained by partial reduction of Mo (VI) to Mo(V) or Mo(IV) species, which has lower partitioning coefficients on an anion exchanger. In nitric acid, this reduction does not appear to occur under these experimental conditions or that the Mo(VI) oxidation state was able to re-establish in the time following irradiation, perhaps due to the oxidation of any reduced Mo species by nitrate ion. A similar reaction would not be expected in the presence of sulfate.

Dissolver for Modified Cintichem process

As party of our efforts to support the International Atomic Energy Agency (IAEA) Coordinated Research Project (CRP) "Developing Techniques for Small-Scale, Indigenous Production of Mo-99 Using Low-Enriched Uranium (LEU) or Neutron Activation (T1.20.18)," we are developing a small-foot-print dissolver for LEU-foils as part of the LEU-modifiedCintichem process. Previous demonstrations done at BATAN (Indonesia) have utilized their existing dissolver that was designed for HEU oxide targets.[23-25] The dissolver (Figure 9) is constructed of Stainless Steel 304 [schedule 40 nominal 3.8 cm (1.5 inch) pipe, 22.9 cm (9 inches) tall with a welded cap bottom], has a volume of 360 cm^3, and rated limits of 900°C and 670 Pa (2000 psig). The top is a flanged lid with an o-ring seal, and a "cross" fitting with a pressure gauge, a plug valve attached to a quick-connect plug, and a plug valve attached to a septum.

The dissolver support sleeve (Figure 9) is a cylinder with an open base and a flange at the top. The sleeve is 22.9 cm (9 inches) tall and 6 cm (2.38 inch) diameter, and is constructed of 1.6 mm (1/16inch)-thick 304 stainless steel. It is used to support the dissolver during heating. The bottom of the sleeve has a 6.4 cm (2.5 inch) tall window cut out to allow hot air to flow in and around the dissolver.

The heating manifold (Figure 9) is a series of Stainless Steel cylinders constructed of 1.6 mm (1/16inch) 304 Stainless Steel designed to support two operations—(1) heating the dissolver during dissolution and holding and heating a filter containing the Mo-ABO precipitate during the Mo-ABO destruction step. At a right angle is a 5.1 cm (2 inch) tube with a 90° bend facing downward. This tube will hold the heat gun used to blow hot air into the manifold. There are two vertical windows cut into the 6.4 cm (2.5 inch) diameter cylinder on opposite sides from each other to allow heat to vent and to view the destruction of the precipitate during processing. When the dissolver support sleeve is in place, the windows are blocked by the sleeve to direct the hot air up and out. When the precipitate filter is in place, the windows direct the hot air out the

sides and away from the upper filter. This design minimizes the footprint of equipment and simplifies the remote handling needed to complete the procedure.

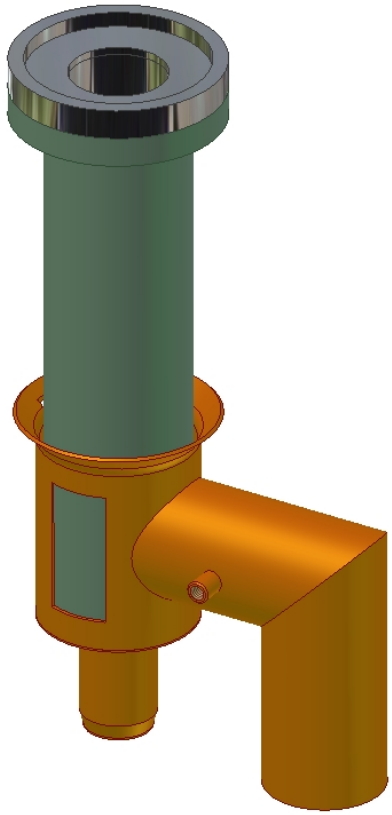

Figure 8. Drawing of the assembled dissolver.

Figure 9. Exploded view of the dissolver. The top piece is the dissolver vessel, the middle piece is the support sleeve and the bottom piece is the heating manifold.

The new dissolver described here will be fabricated at Argonne and tested during a demonstration of the process with full-sized irradiated LEU-foil targets in BATAN's production hot cell facility. When finalized and tested, the design will be made available to our IAEA-CRP partners.

Partnership with CNEA (Argentina)

CNEA has led the development of technology for ^{99}Mo production from LEU. Their production facility has used LEU targets since 2000, utilizing alkaline digestion of LEU-aluminide dispersion plates.[26] In addition, they have provided this process and target to the Australian Nuclear Science and Technology Organization for its ^{99}Mo production. CNEA has also been instrumental in developing and demonstrating the use of LEU-foil targets for alkaline processing. Several LEU-foil targets have been irradiated in the RA-3 reactor and the LEU-modified process has been demonstrated in the Ezeiza ^{99}Mo-production hot cells.

A primary advantages of LEU-foil processing are the significantly smaller volume (~1/5) of base and lower NaOH concentration required to dissolve the LEU foil rather than the HEU target.[27, 2] Argonne has designed and tested a relatively small digester (Figure 10) and small processing rig to recover the uranium precipitate and recovery the Mo on an anion-exchange

column (Figure 11). Demonstration of this equipment and associated ^{99}Mo recovery and purification is planned in the CNEA ^{99}Mo Production Facility in December of 2006. This demonstration will be important for the future of ^{99}Mo production at CNEA by exploring options for future increases in production capacity. The demonstration will also supply important data to other alkaline-side producers (e.g., NTP, Malinkrodt, and IRE) for their conversion analyses.

Figure 10. Photograph of the dissolver during testing.

Figure 11. Photograph of processing stand during testing

Conclusions

The results of this study show that a sorbent column is feasible to separate and purify ^{99}Mo from a LEU nitrate or sulphate based homogeneous reactor fuel solution. Separation and purification of ^{99}Mo from a nitrate based fuel solution would be best achieved using either the Thermoxid sorbents (Isosorb and Radsorb) or the PZC sorbent. Separation and purification of ^{99}Mo from a sulphate based homogeneous reactor solution could only be achieved using Thermoxid sorbents. A new dissolver was designed specifically for the Cintichem Process and will be demonstrated by BATAN (Indonesia). A full demonstration of the alkaline-side LEU-foil process is planned by CNEA (Argentina) for December 2006.

References

1. George F. Vandegrift, Facts and Myths Concerning ^{99}Mo Production with HEU and LEU Targets, Proceedings of the 2005 International Meeting on Reduced Enrichment for Research and Test Reactors, November, 2005, Boston, USA.
2. A. J. Kuperman, Weaker U.S. Export Controls on Bomb-grade Uranium: Causes, Consequenses, and Prospects, Proceedings of the 2005 International Meeting on Reduced Enrichment for Research and Test Reactors, November, 2005, Boston, USA.
3. R.W. Brown, The Radiopharmaceutical Industry's Effort to Migrate Toward Mo-99 Production Utilizing LEU, Proceedings of the 2005 International Meeting on Reduced Enrichment for Research and Test Reactors, November, 2005, Boston, USA.
4. Russell M. Ball, V.A. Pavshook and V.Ye.Khvostionov, Present Status of the Use of LEU in Aqueous Reactors to Produce Mo-99, Proceedings of the International Meeting on Reduced Enrichment for Research and Test Reactors, October, 1998, Sao Paulo, Brazil.

5. McDuffie, H. F., Secoy, C. H., Hochanadel, C. J., McBride, C. J. Aqueous Fuel Systems, In *Reactor Handbook V. 1.* Tripton, C. R. Jr., Ed.; J. Wiley & Sons, 1960.
6. Lane, J. A. Aqueous Fuel Reactors. In *Reactor Handbook, V. 4*. McLain, S., Martens, J. H., Eds.; J. Wiley & Sons, 1964.
7. Ball, R. M. Characteristics of nuclear reactors used for the production of molybdenum-99. In Production *Technologies for Molybdenum-99 and Technetium-99m*. IAEA-TECDOC-1065, IAEA, Vienna, Austria, 1999.
8. Dadachova, K., La Riviere, K., Anderson, P. *J Radioanal. Nucl. Chem*. **1999**, *240*, 935.
9. Stang, L.G. "Manual of Isotope Production Processes in Use at Brookhaven National Laboratory," BNL 864 (T-347), 1964.
10. Boyd, R. E. *Radiochim. Acta,* **1982**, *30*, 123.
11. Betenekov, N. D., Denisov, E. I., Nedobukh, T. A. Sharygin, L. M. United States Patent 6,337,055 (Jan 8, 2002).
12. Ponomarev-Stepnoy, N. N.., Pavshok, V. A., Bebikh, G. F., Khvostionov, V. Y., Trukhlyaev, P. S., Shvetsov, I. K. United States Patent 5,910,971 (June 8, 1999).
13. Ponomarev-Stepnoy, N. N.., Pavshok, V. A., Bebikh, G. F., Khvostionov, V. Y., Trukhlyaev, P. S., Shvetsov, I. K., Vandysh, Y. L. United States Patent 5,962,597 (Oct. 5, 1999).
14. Koehly, G., Madic, C. United States Patent 4,701,308 (Oct. 20, 1987).
15. Arino, H., Kramer, H. H., McGovern, J. J., Thorton, A. K. United States Patent 3,799, 833 (Mar. 26, 1974)
16. Arino, H., Kramer, H. H. *Int. J. Appl. Rad. Isot.* **1978**, *29*, 97.
17. Cheng, W. L., Lee, C. S., Chen, C. C., Wang, Y. M., Ting, G. *Appl. Radiat. Isot.,* **1989**, *40*, 315.
18. Sivaramakriahnan, C. K. BARC-847, 1976.
19. Horwitz, E. P.; Chiarizia, R.; Dietz, M. L. *Solvent Extr. Ion Exch.* **1992,** *10,* 313.
20. Horwitz, E. P.; Chiarizia, R.; Diamond, H.; Gatrone, R. C.; Alexandratos, S. D.; Trochimzuk, A. W.; Creek, D. W. *Solvent Extr. Ion Exch.* **1993**, *11,* 943.
21. Horwitz, E. P.; Dietz, M. L.; Chiarizia, R.; Diamond, H.; Maxwell S.L.; Nelson, M. R. *Anal. Chim. Acta* **1995**, *310,* 63.
22. Chiarizia, R.; Horwitz, E. P.; Alexandratos, S. D. *Solvent Extr. Ion Exch.* 2004, *12,* 211.
23. Mutalib, A.; Purwati, B.; Adang, H. G. ; Hotman, L. ; Moeridoen; Kadarisman ; Sukmana, A.; Sriyono, Suripto, A.; Nasution, H.; Amin, D. L.; Basiran, A.; Gogo, A.; Sunaryadi, D.; Taryo, T.; Vandegrift, G. F.; Hofman, G.; Conner, C.; Sedlet, J.; Walker, D.; Leonard, R. A.; Wood, E. L.; Woencek, T. C.; and Snelgrove, J. L. Full-scale Demonstration of the Cintichem Process for the production of 99Mo using a Low Enriched Target. Proceedings of the International Meeting on Reduced Enrichment for Research and Test Reactors, October, 1998, Sao Paulo, Brazil.
24. Vandegrift, G. F.; Hofman, G. L.; Conner, C.; Snelgrove, J. L. ; Mutalib, A.; Purwati, B.; Adang, H. G. ; Hotman, L. ; Kadarisman ; Sukmana, A.; Dicky, T. J.; Suripto, A.; Lutfi, D.; Amin, D. L.; Basiran, A.; Gogo, A.; Sarwani; and Taryo, T. Demonstration of ^{99}Mo Production Using LEU Metal Foil Targets in the Cintichem Process, Proceedings of the International Meeting on Reduced Enrichment for Research and Test Reactors, October, 1999, Budapest, Hungary.
25. Conner, C.; Sedlet, J.; Weincek, T. C.; McGann, D. J.; Hofman, G. L.; Vandegrift, G. F.; Snelgrove, J. L. ; Mutalib, A.; Gunawan, A. H.; Adang, H. G. ; Lubis, H.; Wisnukaton, K.; Kadarisman ; Sukmana, A.; Sriyono; Purwadi, B; Jatmiko, D. T.; Suripto, A.; Amin, D. L.; Basiran, A.; Martoyo; Sarwani; and Taryo, T. Production of ^{99}Mo from LEU Targets—Acid Side Processing. Proceedings of the International Meeting on Reduced Enrichment for Research and Test Reactors, October, 2000, Las Vegas, USA.

26. Pablo R. Cristini, Héctor J. Cols, Ricardo Bavaro, Marcelo Bronca, Roberto Centurión, Daniel Cestau Production of Mo-99 from Low Enriched Uranium Targets, Proceedings of the International Meeting on Reduced Enrichment for Research and Test Reactors, September, 2002, Bariloche, Argentina.
27. A.V. Gelis, G. F. Vandegrift, , S. B. Aase, A. J. Bakel, J. R. Falkenberg, M. C. Regalbuto, and K. J. Quigley, "ANL Progress in Developing a Target and Process for Mo-99 Production: Cooperation with CNEA," Proceedings of the 2003 International Meeting on Reduced Enrichment for Research and Test Reactors, October, 2003, Chicago, IL, USA.
28. A. V. Gelis, K. J. Quigley, S. B. Aase, A. J. Bakel, A. Leyva, M. C. Regalbuto, and G. F. Vandegrift, ANL Progress On the Cooperation with CNEA for the Mo-99 Production: Base-Side Digestion Process, Proceedings of the 2003 International Meeting on Reduced Enrichment for Research and Test Reactors, November, 2004, Vienna, Austria.